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Acoustic metamaterials through a microfluidic, bottom-up approach: Toward highly attenuating, negative effective density materials

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14. ABSTRACT
We report the achievement of highly monodisperse emulsions exhibiting about ten acoustic Mie resonances. Thanks to robotics (microfluidics), the effective acoustic properties of such strongly scattering media can be precisely targeted by means of the production of calibrated (random) liquid-droplets. Ultrasonic experiments are compared, with an excellent quantitative agreement, to theoretical predictions derived within the framework of the independent scattering approximation. The dependence of the sound speed and of the acoustic attenuation on both the size and the volume fraction of droplets is quantitatively examined for dilute and more concentrated emulsions.

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Final report on the EOARD project FA8655-11-M-4006

"Acoustic metamaterials through a microfluidic, bottom-up approach: Toward highly attenuating, negative effective density materials"

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1) <u>Our approaches</u>

Our initial idea was to build metamaterials with specific properties such as large attenuation capacities in a targeted field of frequencies. If much work has been up to now performed using centimetre-sized handmade mechanical resonators adapted to audible frequencies [1-3], fewer attempts have been made on smaller micro-resonators, in the 100 µm range, i.e. for ultrasonic frequencies. Following this path, we recently proposed two different microfluidic approaches to get such micro-resonators:

1. The "core-shell" approach
2. The "resonant emulsion "approach

In the first one, we aimed at fabricating core-shell objects composed of a hard core exhibiting a high density contrast with a surrounding elastic soft shell, the resulting core-shell being finally embedded in a relatively hard solid matrix. Predictions [4] and earlier experimental results [1] show that, at a given frequency, the core-shell particle behaves as an oscillator which resonates with the incoming acoustic wave. The expected resonance occurs in that case at low frequency $f_{res} \sim 1/a$.

In the second approach, one aims at fabricating a material in which the incoming wave will exhibit a "Mie" type of resonance with the inclusions. In such case, the expected cavity resonance is due to the sound velocity contrast between the inclusions and the surrounding matrix. Resonance occurs at a frequency $f_{res} \sim ((n+1)/2)\ .(c/a)$ where c and a are respectively the sound velocity in the inclusion and its characteristic size and n, an integer, the order of the resonance. Of course, significant resonating effects may only occur if the sound contrast between the inclusion and its surrounding medium is large enough, as noted by Li *et al* [5]. Earlier experimental works showed that interesting resonating behaviour can be obtained with air bubbles [6]. However, air bubbles are hardly stable in time and we proposed an alternative experimental route to achieve materials containing only liquids.

2) <u>What has been done from October – May 2012: the resonant emulsions approach</u>

For the sake of efficiency and considering the time schedule of this USAF project, we decided to concentrate our efforts on the second approach which consists in fabricating a soft material composed of oil emulsion droplets (made with of fluorinated oil) suspended in a liquid water-based gel. We indeed identified that fluorinated oil (FC40) possesses a rather low sound speed for a pure liquid (c = 640 m/s) and performed some materials composed of micrometric oil droplets suspended in a Bingham fluid (a fluid that does behave as a solid under a given threshold stress, in our case a solution of polyacrylic acid (carbopol) in water) that has a sound speed of ~ 1500 m/s. An example of such a material obtained through a microfluidic approach is shown on figure 1. In this approach, one can vary the average size and droplets volume fraction in the material.

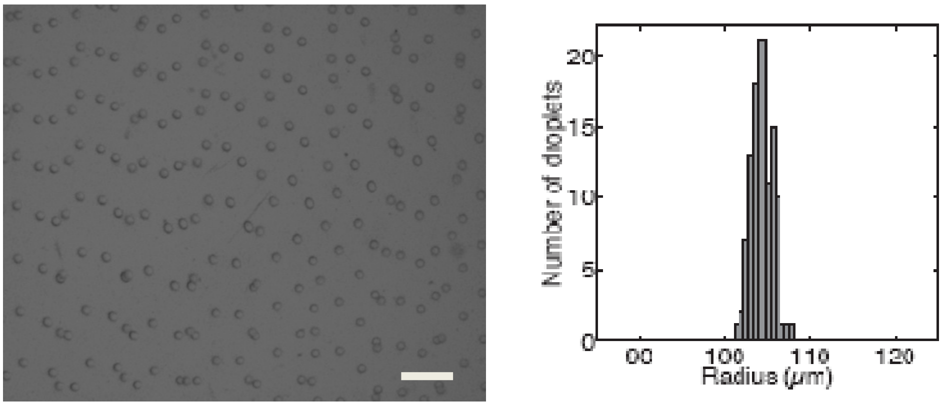

Figure 1:
a. Optical microscopy picture of FC40 oil droplets dispersed in a carbopol/water mixture (scale bar: 1mm); *b.* Corresponding droplets size distribution

Within the frame of this USAF project, the hired postdoc (Benoit Mascaro) developed the experimental set-up required for the acoustic measurements inside the fluid (see figure 2). On this picture a 2.5 cm-thick plastic cell containing the material is introduced between an acoustic emitter and a receptor and both the acoustic attenuation and the phase velocity are measured as a function of the wave frequency f that is, in that case, varied between 1 and 10 MHz.

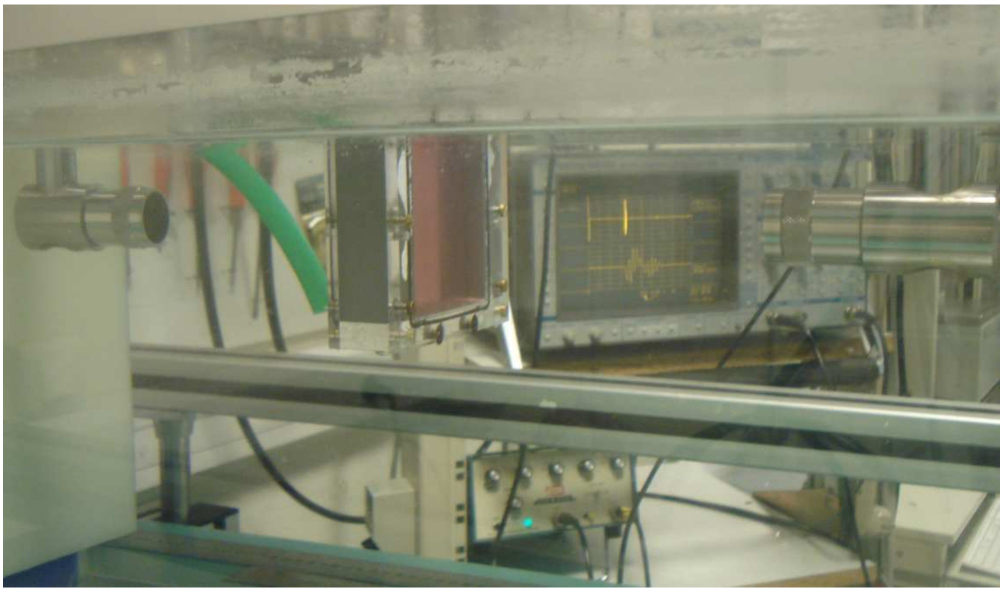

Figure 2:
Picture of the acoustic experimental set-up. The acoustic transducers are both immersed in water.

On figure 3, we report the measurements performed in a material containing ϕ_V = 0.23 vol % of droplets with average diameters 2R = 104 +/- 1 µm. The measure clearly evidences the resonating behavior of the system. The different attenuation peaks corresponds to different orders n (the size of the inclusion being equal to an integer number of half wavelengths). The results could be fitted using the Independent Scattering Approximation (ISA) [7] that considers no interactions between the acoustic waves scattered by the inclusions. Leaving $2R$ and ϕ_V as the only free parameters of the fit, one gets an excellent agreement (solid lines on figure 3) with the data with $2R$ = 103.5 µm and ϕ_V = 0.22%, i.e some values very close to the experimental ones. The agreement is even better when the experimental droplets polydispersity is taken into account in the calculation (see dashed line in figure 3).

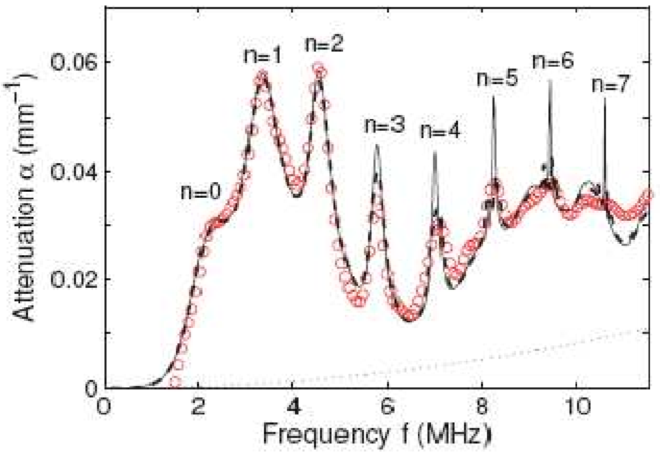

Figure 3:

Evolution of the attenuation (mm^{-1}) as a function of the wave frequency *f*. Solid line: fitting of the data to the ISA model using $2R$ = 103.5 µm and ϕ_V = 0.22% and considering no polydispersity; Dashed line: fitting of the data to the ISA model and taking into account the sample polydispersity.

3) What has been done since May 2012: Impact of polydispersity on the resonant behavior

Since the last report (May 2012), we have investigated the impact of polydispersity on multipolar resonant scattering. To achieve samples with controlled polydispersities, we performed droplets with slightly different diameters using the process described in section 2 and mix them together. This way, we could obtain targeted particle size distribution (PSD) as presented in figure 4. The PSD of each sample was characterized by optical measurements on about a hundred fluorinated-oil droplets. The mean radii <a> were about a hundred microns and the size-polydispersities $\sigma/\langle a\rangle$ ranged from 1% to 13% (see Fig. 4). The latter parameter can also be viewed as a quality factor $Q_{PSD} = \langle a\rangle/\sigma$. The volume fraction ϕ_V of oil-droplets was intentionally fixed to values as low as 1% in order to facilitate the experiments. The acoustical measurements of the attenuation coefficient and phase velocity for each sample are displayed versus the adimensional frequency as illustrated in Fig. 4. The mean radius of the oil droplets dispersed in each sample being not identical, the representation versus the reduced frequency *ka* is the most appropriate for a direct comparison between the resonant scattering of the different emulsions. By using an inversion technique based on a least squares optimization a best fit procedure permits the evaluation of the parameters characterizing the PSD: the mean radius $\langle a\rangle$, the size polydispersity $\sigma/\langle a\rangle$ and the volume fraction ϕ_V, from the acoustic measurements. The values determined by optical and acoustical measurements summarized in Tab. I have been found to be very close. In particular, the size polydispersities of samples are well-recovered by acoustical measurements. Thus, the resonant regime could provide an accurate method to characterize the complete PSD of particulate mixtures such as emulsions or suspensions, usually studied in the long wavelength regime.

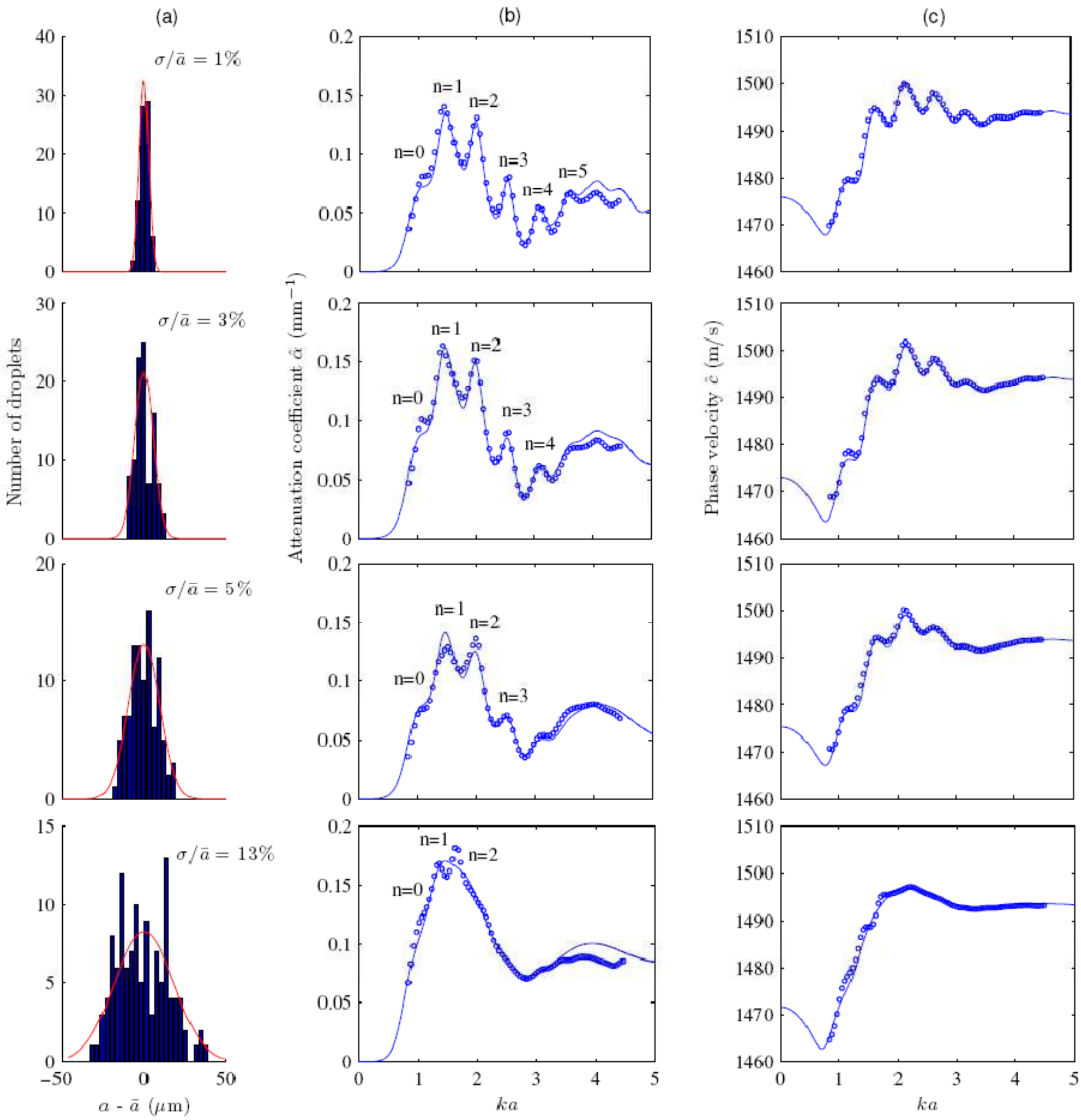

Figure 4:

Optical and acoustical properties of the emulsion samples with increasing polydispersity. (a) centered droplet-radius histograms obtained from optical microscopy measurements. (b) attenuation coefficient and (c) phase velocity: (o) acoustical measurement and (solid line) computations from the optimal polydisperse gaussian distribution (best fit of the experimental data to the model).

Sample number	$\langle a \rangle_{opt}$ (µm)	$(\sigma/\langle a \rangle)_{opt}$ (%)	$\phi_{V,opt}$ (%)	$\langle a \rangle_{ac}$ (µm)	$(\sigma/\langle a \rangle)_{ac}$ (%)	$\phi_{V,ac}$ (%)
1	185	1.4	≈ 1	181	2.41	0.95
2	174	3.1	≈ 1	173	3.6	1.1
3	172	4.9	≈ 1	170	4.82	0.98
4	128	12.7	≈ 1	142	13.4	1.19

Table I:

Compared values of the size, size distribution and volume fraction deduced from optical observations and acoustic characterization

Fig. 4 also shows that the scattering resonant features progressively disappear as the size polydispersity increases. For example, the attenuation peaks are clearly observable until n = 5 for the 1%-polydisperse emulsion (sample 1) while the fundamental resonances of the first three modes (n = 0; 1; 2) are hardly identifiable for a 13%-polydisperse emulsion (sample 4). Tab. II makes an inventory of the observable (o) or non observable (X) attenuation peaks for each sample. The narrowness of the resonance peaks is characterized by the quality factor $Q_{res}= f_{res}/\Delta f_{res}$, of which values are reminded in Tab. II. Thus, the two parameters Q_{res} and Q_{PSD} can be quantitatively compared for all resonances occurring in each sample. Tab. II shows then that Q_{PSD} must be higher than Q_{res} in order to clearly observe a distinguishable attenuation peak. All resonances, for which the quality factor Q_{res} is higher than Q_{PSD}, do not exhibit prominent features in the attenuation and phase velocity spectra.

	n = 3 $Q_{res} \approx 10$	**n = 4** $Q_{res} \approx 24$	**n = 5** $Q_{res} \approx 56$	**n = 6** $Q_{res} \approx 140$
$\sigma/\langle a \rangle$ = 1 % **Q_{PSD} = 100**	o	o	o	X
$\sigma/\langle a \rangle$ = 3 % **Q_{PSD} = 33**	o	o	X	X
$\sigma/\langle a \rangle$ = 5 % **Q_{PSD} = 20**	o	X	X	X
$\sigma/\langle a \rangle$ = 13 % **Q_{PSD} = 7.5**	X	X	X	X

Table II:

Synthesis of observable (o) or non-observable (X) fundamental acoustic resonances of several modes ($3 \leq n \leq 6$) occurring in fluorinated oil droplet emulsions with different size polydispersities $\sigma/\langle a \rangle$ ranging from 1% to 13%. For comparison, the quality factor Q_{res} of these fundamental resonances are mentioned for each mode as well as the inverse of the size polydispersity, $Q_{PSD} = \langle a \rangle / \sigma$, which characterizes the particle-size distribution of the samples.

This simple approach thus provides a reasonable way to evaluate the impact of polydispersity on the resonance amplitude.

4) What we are currently doing at the moment: tunable resonant materials

We now aim at testing the impact of the sample volume fraction on the resonances. The used ISA is very well adapted to the case of low volume fractions of scattering inclusions that were studied up to now. We now work on the introduction of higher density number of inclusions in order to enter the regime of multiple scattering for which the interaction between scattered acoustic waves is taken into account. This will permit to explore the possibility of using other models to describe our data.

Another part of our work also consists in the fabrication of new experimental systems. As shown by now, the frequency positions of the attenuation peaks strongly depend on the size and the shape of the inclusions. We recently obtained from USAF a follow-up of this project that will be the introduction of controlled amount of iron oxide particle inside the oil droplets. As a consequence, when submitted to an external magnetic field, the droplets will get deformed and the peak frequencies should be moved to different values.

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[3] Z. Yang *et al.*, *Applied Physics Letters* **96**, 041906 (2010)

[4] Z. Liu *et al.*, *Phys. Rev. B* **71**: 014103 (2005)

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[7] P. Sheng, *Introduction to Wave Scattering, Localization, and Mesoscopic Phenomena* (Springer, Heidelberg), 2006

6) Participation to conferences

B. Mascaro: Oral presentation at Anglo-French Physical Acoustics Conference, 18-20 January 2012, Brighton, UK

T. Brunet: Poster at the international symposium on Ultrasound in the Control of Industrial Processes, 18-20 April 2012, Madrid, Spain

T. Brunet: Oral presentation at Acoustics 2012, 23-27 April 2012, Nantes, France

O. Mondain-Monval: Invited talk at the “International Conference on Materials Science and Technology”, 10-14 June 2012, Kottayam, India

O. Mondain-Monval: Invited seminar for the *Dr R A Mashelkar Endowment Lecture Series in Advanced Materials*, June 8th 2012, National Chemical Laboratory, Pune, India

O. Mondain-Monval: Invited seminar, June 15th 2012, Indira Ghandi Center for Atomic Research, Kalpakkam, India

7) <u>Publications</u>

”Sharp acoustic multipolar-resonances in highly monodisperse emulsions”, Thomas Brunet, Simon Raffy, Benoit Mascaro, Jacques Leng, Régis Wunenburger, Olivier Mondain-Monval, Olivier Poncelet and Christophe Aristégui, **Applied Physics Letters 101**, 011913 (2012)

“Impact of polydispersity on multipolar resonant scattering in emulsions”, Benoit Mascaro, Thomas Brunet, Olivier Poncelet, Christophe Aristégui, Simon Raffy, Olivier Mondain-Monval, Jacques Leng, submitted to the **Journal of The Acoustic Society of America**